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6. AUTHOR(S)
John W. Daily

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Department of Mechanical Engineering
University of California, Berkeley, CA 94720

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INTRODUCTION

The study of reacting gas flows has traditionally been limited by a lack of experimental data. The measurement of temperatures and species concentrations in complex flames with sufficient spatial and time resolution would be especially desirable. Under AFOSR Grant 77-3357 we have been investigating the application of Laser Induced FLuorescence Spectroscopy (LIFS) to measurement of species concentrations and temperatures in flames. This report describes progress made during the grant period 1 May 1977 through 30 April 1981.

During the past four years we have concentrated on addressing questions which arise if one wishes to measure the concentration of atomic and diatomic species and temperature in turbulent flames.

The LIFS method consists of illuminating the gas with a laser tuned to an absorption line of the species of interest. The species is excited and fluoresces. The fluorescence is observed at 90° to the laser beam, the volume formed by the intersection of the laser beam and the collection optics defining the spatial resolution. If a relationship between the number density can be found, then the fluorescence is a measure of the total number density. Furthermore, if a measurement can be made with a single pulse, then turbulent measurements are possible. If, in addition, part or all of the fluorescence spectrum is obtained then it is also possible to recover the flame temperature.

Virtually all of our work has been reported in the open, refeered literature. In the following we abstract each paper with its reference.

Pulsed Resonance Spectroscopy Applied to Turbulent Combustion Flows

John W. Daily, Applied Optics, 15, 955 (1976)

The application of pulsed resonance spectroscopy to the measurement of species concentrations in chemically reacting turbulent flows is discussed. The theory of such measurements is developed. An uncertainty analysis is performed and the results applied to typical combustion conditions. Experimental aspects are discussed. The method shows a significant theoretical improvement in accuracy over Raman and Rayleigh scattering techniques, and no significant barriers appear to block its development.

Saturation Effects in Laser Induced Fluorescence Spectroscopy

John W. Daily, Applied Optics, 16, 568 (1977)

Laser based spectroscopic diagnostic tools offer the possibility of spatially and temporally resolved measurements of species concentrations in complex reacting gas flows of engineering interest. The major problem associated with such measurements is the effect of quenching reactions on the fluorescence signal. To overcome this difficulty operating in the saturation mode is proposed. For suitable systems the fluorescence signal is then no longer a function of quenching rates or laser power. Very low detectability limits appear possible.

Use of Rate Equations to Describe Laser Excitation in Flames

John W. Daily, Applied Optics, 16, 2322 (1977)

In describing the exictation of atomic and molecular species with lasers in spectroscopic applications, only the density matrix formulation is exactly correct. Many workers, however, have used the conventional rate equation formulation. The range of application of the conventional rate equations is examined and, for flames, shown to be valid for sufficiently slow laser pulse rise times under single-mode excitation and for certain special cases of multimode excitation.

Laser-Induced Fluorescence Measurement of Sodium in Flames

John W. Daily and Calvin Chin, Combustion Flame, 33, 47-53 (1978)

In the present work the use of Saturated Laser-Induced Fluorescence Spectroscopy to measure sodium-atom concentrations in flames is described. A flashlamp pumped-dye laser is used to excite the sodium, and the fluorescence signal is observed at 90° forming a focal volume about 27 μm by 1 mm. A linear curve of growth is obtained in an atmospheric flame for concentration in the range 0.008-0.2 PPM.

Saturation of Fluorescence in Flames with a Gaussian Laser Beam

John W. Daily, Applied Optics, 17, 225 (1978)

The method of saturated fluorescence for measuring species concentrations in flames is usually performed with laser beams that do not provide a constant intensity distribution across the focal volume. Because of the intensity distribution across the beam, the fluorescence signal does not depend on laser power or intensity in the same manner as for uniform illumination. This leads to anomalous apparent saturation intensities. In the following, the effect is considered for atomic fluorescence. Relations for the fluorescence signal under two common excitation geometries are derived and uncertainty relations used to consider the benefits of high laser intensity.

Detectability Limit and Uncertainty Considerations for Laser Induced Fluorescence Spectroscopy in Flames

John W. Daily, Applied Optics, 17, 1610 (1978)

Uncertainty relations are derived for fluorescence caused by laser excitation in flames. The effect of statistical and systematic uncertainities on the detectability limits for excited states are examined. Fluorescence trapping is discussed and shown to establish an upper limit on the number density that can be measured. Methods for minimizing the trapping effect are discussed.

Coherent Optical Transient Spectroscopy in Flames

John W. Daily, Applied Optics, 18, 360 (1979)

The use of coherent optical transient spectroscopy to study collisional processes in flames has been considered. A short discussion of the density matrix equations is used to illustrate transient phenomena of possible interest. Characteristic times that arise for such phenomena are calculated for a variety of flame conditions, and it is concluded that both dephasing and energy decay rates for electronic and ir transitions in selected species may be measured. Available excitation sources and transient measurement techniques are examined and found suitable for making such measurements.

Near-Resonant Rayleigh Scattering and Atomic Flame Fluorescence Spectroscopy

Calvin Chan and John W. Daily
Quant. Spectrosc. Radiat. Transfer, 21, 527-531 (1979).

Near-resonant Rayleigh scattering and collision-induced resonance fluorescence of sodium have been observed in a combustion environment at atmospheric pressure. Sodium atoms in the equilibrium region of a flat-flame burner were excited by a pulsed-dye laser. The behavior of the two signals as the laser was detuned from the resonance line was studied and was found to agree with theoretical predictions. The results have important implications for atomic flame-fluorescence spectroscopy in that trapping is avoided at large atom number densities.

Laser Excitation Dynamics of OH in Flames

Calvin Chan and John W. Daily, Applied Optics, 19, 1357 (1980)

The response of OH in flames to laser excitation is studied in some detail. The population balance equations are solved numerically using an empirical model for the rotational relaxation rates. The empirical model parameters are fit to experimental spectra using a linear regression procedure and the resulting description of OH behavior is shown to be satisfactory to within the precision of the data. The model is then used to recover branching ratios for a number of flame conditions.

Measurement of Temperature in Flames Using Laser Induced Fluorescence Spectroscopy of OH

Calvin Chan and John W. Daily, Applied Optics, 19, 1963 (1980)

A technique for measuring translational flame temperature utilizing the laser induced fluorescence spectrum of OH is demonstrated. The method is based on matching the observed spectrum to a numerical model in which the detailed balance temperature is a parameter. The precision of the method exceeds that of the line reversal technique. Accuracy is limited by the calibration source and the validity of the numerical rotational relaxation rate model.